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# Different Sub- $T_g$ Relaxation Patterns in Metallic Glasses far from Equilibrium

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We investigate the sub- $T_g$  relaxation patterns (RPs) in CuZrAl and LaAlNi glass ribbons (GRs) by using the hyperquenching-sub- $T_g$  annealing-calorimetric approach.<sup>1-3</sup> Different from the monotonic relaxation pattern observed in rare-earth based metallic GRs, the abnormal three-steplike sub- $T_g$  relaxation pattern has been observed in CuZrAl ternary glasses. It has been found that the presence of this three-steplike sub- $T_g$  RPs of CuZrAl closely relates to the fictive temperature  $T_f$  where the supercooled liquids are frozen in during cooling. Only in a certain range of  $T_f$  (or cooling rate for fabrication), this thermodynamic anomaly can be observed. Accompanied with this thermodynamic anomaly, we have observed a three-stage evolution of structures with the increase of annealing degree on CuZrAl GRs. It indicates a dramatic change of the MRO clusters around 1.3  $T_g$  upon cooling in supercooled liquids, which is due to the competitions among the MRO clusters composed of different locally ordering configuration.<sup>3,4</sup> This accords well with the models based on the competitions between the low-temperature and high-temperature clusters used in the terms of the fragile-to-strong (F-S) transition. In order to further clarify the generality and the origin of this remarkable abnormal thermodynamic phenomenon, the sub- $T_g$  relaxation patterns (RPs) was also detected in other Cu-based metallic GRs. It has been found that this abnormal RPs can be observed in binary CuZr and ternary CuZrAl glasses, rather in quaternary CuZrAlNi. It shows that the anomaly of sub- $T_g$  relaxation pattern could not be simply attributed to the large difference in the enthalpy of mixing between different elements in alloys. By using a high-temperature torsional oscillating viscometer, the existence of the liquid-liquid phase transition (LLPT) depicted by anomalous viscosity drop during cooling in superheated liquids is also been observed in CuZr and CuZrAl melts. In accordance with this dynamic anomaly well above the liquidus temperature, the thermodynamic evidence in these alloys has also been detected above  $T_l$ . A close link between the abnormal three-steplike sub- $T_g$  relaxation pattern in GRs and the LLPT in their corresponding melts has been discovered. This work helps to better understand the complex structural evolution from superheated to glassy solids approaching  $T_g$ .

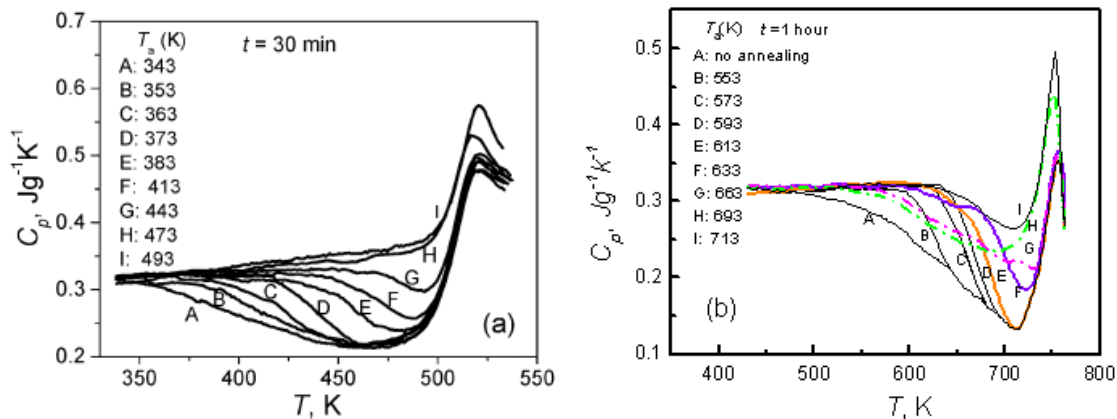


Fig. 1 Sub- $T_g$  relaxation pattern of the hyperquenched GRs (a)  $\text{La}_{55}\text{Al}_{25}\text{Ni}_{20}$ ; (b)  $\text{Cu}_{45}\text{Zr}_{45}\text{Al}_{10}$ . All the  $C_p$  curves were obtained at a heating rate of 20 K/min.  $T_g$  points to the annealing temperature and  $t$  points to the annealing time. The curves G and H in (b) reflect the non-monotonic relaxation pattern of CuZrAl glass ribbons.

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